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Interpretation of holographic phase shifts from polar interfaces, ferroelectric nanostructures and charged dielectric materials

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Electron holography experiments carried out on polar materials have proven extremely challenging [1,2]: contrary to ferromagnets, where the electron beam is affected by the magnetization, polarization alone is invisible to phase-sensitive techniques. The origin of this fundamental difference can be traced back to the Lorentz force: magnetic deflections are proportional to the B-field, of which M is an integral part, while electric deflections are proportional to the E-field; an electric field might as well be generated by bound polarization charges if present, but with a non-linear and, in a sense, non-local relationship as polarization charges may be very far away from the illuminated region of the sample. Moreover, the absence of magnetic monopoles combined with quantization of angular momentum, which applies only to spins and not to electric dipoles, results in screening/depolarizing effects manifesting themselves very differently in magnetic and electric polar materials.

In nanoparticles made of materials that are ferroelectric in bulk form, the polarization is suppressed by its own depolarization field [3]. To preserve P below a certain critical size, screening of the depolarization field must occur. Screening can be internal --redistribution of mobile charges in metallic or oxygen-vacant insulating ferroelectrics-- or external in case of embedment in a dielectric or metallic matrix. However, if the screening is 100% effective the internal electric field vanishes entirely, giving rise to no observable holographic signal. A trade-off needs to be found between screening capacity and magnitude of the resulting polarization that maximizes the residual internal field producing observable phase shifts. This trade-off will be discussed, highlighting realistic and achievable experimental conditions that may facilitate the success of holography experiments on ferroelectric nanostructures.

An omnipresent complication when interpreting holographic signals from nanoparticles originates from beam-induced charging. A poorly conductive object tends to charge positively upon irradiation due to secondary electron generation. When the object is supported by a conductive substrate, image charge effects will produce an overall dipole field from a charge object, i.e. with the same expected character as the stray field from polarization. As a result, separating charging and polarization is extremely challenging, and can be accomplished only by proper modelling and careful measurement strategies.

In case of polar interfaces [4], for example a ferroelectric layer sandwiched in between dielectrics, or heterostructures involving charge-layered oxide materials such as LaAlO₃ in contact with doped or undoped SrTiO₃ where a 2D electron gas may emerge, determining whether or not we can expect a phase shift is a difficult task. In addition to self-depolarization, particularly strong in parallel-plate-capacitor situations, we need to handle the band structure relaxation bringing extra mobile charges into the picture: charge transfer may occur even with insulators, if band bending is so severe that the chemical potential dips into their valence band. A test scenario involving two ferroelectrics with different bulk polarizations, with and without chemical potential shift will be presented to illustrate the complexity of the problem, and to provide an initial framework for the interpretation of holographic signals from polar interfaces.

References:

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